

Docket No.: 241959US0

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:

GROUP: 1621

Masaki TAKAI, et al.

SERIAL NO: 10/649,767

EXAMINER: KEYS, R. A.

FILED: August 28, 2003

FOR: METHOD FOR PRODUCING
ALLYL COMPOUND

DECLARATION UNDER 37 C.F.R. § 1.132

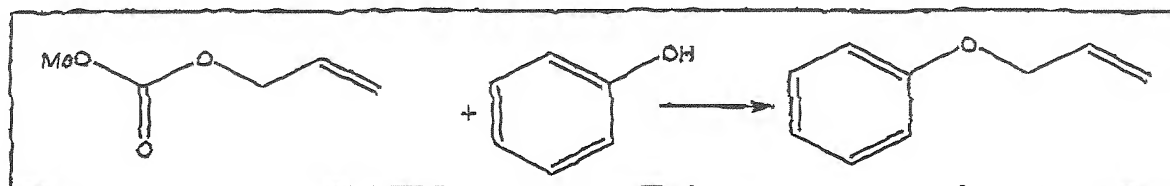
COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

Sir:

Now comes Masaki Takai who deposes and states that:

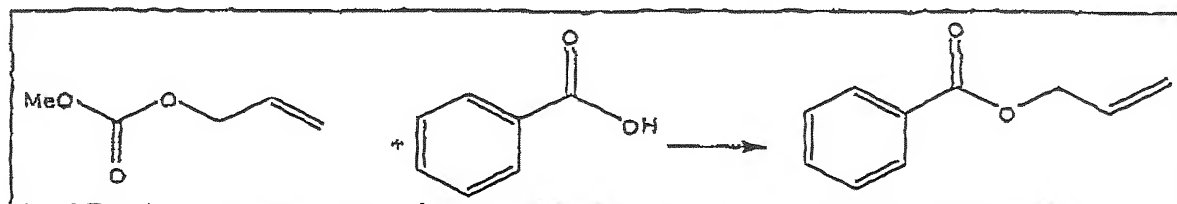
1. I am a graduate of Tokyo University and received my master degree in the year 1985.
2. I have been employed by Mitsubishi Chemical Corporation for 22 years as a researcher in the field of homogeneous catalysis and organic synthesis.
3. The following experiments were carried out by me or under my direct supervision and control.
4. The Examples in the specification show the comparative yields obtained when triisopropyl phosphite and triethyl phosphite were used as the monodentate phosphite.
5. The effects of varying the alkyl chain length in the compound of formula (I) on yield of allyl products are shown in the specification and by the experimental data below.

6. The experimental data in this section refer to the following chemical reaction which produces allyl phenyl ether.



Formula (I) compound		Yield (allyl phenyl ether)
Triisopropyl phosphite	Example 1	97%
Triethyl phosphite	Example 2	50
Tributyl phosphite	Declaration	66
Tris (2-ethylhexyl) phosphite	Declaration	58
Trisodecyl phosphite	Declaration	45
Triphenyl phosphite	Comp. Ex. 2	3
Tris (2,4-di- <i>t</i> -butylphenyl) phosphite	Comp. Ex. 3	0
DPPB (bidentate phosphine)	Comp. Ex. 1	4

7. The experimental data in this section refer to the following chemical reaction which produces allyl benzoate.



Formula (I) compound		Yield (allyl benzoate)
Triisopropyl phosphite	Example 4	60%
Tributyl phosphite	Declaration	59
Tris (2-ethylhexyl) phosphite	Declaration	65
Trisodecyl phosphite	Declaration	54
Triphenyl phosphite	Comp. Ex. 9	21
DPPB (bidentate phosphine)	Comp. Ex. 8	1

8. The new experimental data described above were obtained as follows.

Allylmethyl carbonate + phenol → allyl phenyl ether

Allylation was carried out with an alkyl phosphite as a ligand by using allyl methyl carbonate as an allyl starting material compound and phenol as an oxygen nucleophilic agent, in accordance with the examples of JP-A-2004-107337 which is a laid-open publication of Japanese Patent Application No. 2003-306075 which is an Internal Priority Application claiming the priority of the original Japanese Application No. 2002-251620. 0.0147 g (0.0160mmol) of trisbenzylideneacetone palladium as a transition metal compound was put in an Schlenk flask, and the inside of the flask was displaced with argon. Tributyl phosphite as a trialkyl type monodentate phosphite was added thereto in an amount of 8 equivalents to palladium, and then 2.0 ml of a commercially available dried tetrahydrofuran was added thereto, followed by stirring at room temperature to obtain a catalyst solution having a palladium concentration of 16.0mmol/l. Another Schlenk flask for reaction was displaced with argon, and then 5.0 ml of a tetrahydrofuran solution containing 0.192 g (1.65 mmol) of allyl methyl carbonate and 0.274g (2.91mmol) of phenol was added thereto in an argon atmosphere. Then, 20.0 μ l of the above catalyst solution was added with a microsyringe, followed by heating at 60°C for reaction. After 30 minutes reaction, the composition of the solution was analyzed with a gas chromatography to determine the yield of allyl phenyl ether. The reaction was carried out under the same conditions provided that tris(2-ethylhexyl) phosphite or triisodecyl phosphite was used as a ligand.

Allyl acetate + benzoic acid → Allyl benzoate

Allylation was carried out with an alkyl phosphite as a ligand by using allyl acetate as an allyl starting material compound and benzoic acid as an oxygen nucleophilic agent, in accordance with the examples of JP-A-2004-107337. 0.0150 g(0.0164 mmol) of trisbenzylideneacetone palladium as a transition metal compound was put in a Schlenk flask,

and the inside of the flask was displaced with argon. Tributyl phosphite as a trialkyl type monodentate phosphite was added thereto in an amount of 8 equivalents to palladium, and then 2.0 ml of a commercially available dried tetrahydrofuran was added thereto, followed by stirring at room temperature to obtain a catalyst solution having a palladium concentration of 16.4 mmol/l. Another Schlenk flask for reaction was displaced with argon, and then 4.0 ml of a tetrahydrofuran solution containing 0.123 g (1.23mmol) of allyl acetate and 0.304 g (2.49 mmol) of benzoic acid was added in an argon atmosphere. Then, 60.0 μ l of the above catalyst solution was added with a microsyringe, followed by heating at 60°C for reaction. After 30 minutes reaction, the composition of the solution was analyzed with a gas chromatography to determine the yield of allyl benzoate. The reaction was carried out under the same conditions provided that tris(2-ethylhexyl) phosphite or triisodecyl phosphite was used as a ligand.

9. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

10. Further deponent saith not.

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Signature

Masaki Takai

Date

Sep. 21, 2007